

Broad-band spectral control of single photon sources using a nonlinear photonic crystal cavity

Murray W. McCutcheon*,¹ Darrick E. Chang*,² Yinan Zhang,¹ Mikhail D. Lukin,³ and Marko Lončar¹

¹*School of Engineering and Applied Sciences, Harvard University, Cambridge, MA 02138**

²*Institute for Quantum Information and Center for the Physics of Information,
California Institute of Technology, Pasadena, CA 91125*

³*Physics Department, Harvard University, Cambridge, MA 02138*

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Motivated by developments in quantum information science, much recent effort has been directed toward coupling individual quantum emitters to optical microcavities. Such systems can be used to produce single photons on demand, enable nonlinear optical switching at a single photon level, and implement functional nodes of a quantum network, where the emitters serve as processing nodes and photons are used for long-distance quantum communication. For many of these practical applications, it is important to develop techniques that allow one to generate outgoing single photons of desired frequency and bandwidth, enabling hybrid networks connecting different types of emitters and long-distance transmission over telecommunications wavelengths. Here, we propose a novel approach that makes use of a nonlinear optical resonator, in which the single photon originating from the atom-like emitter is directly converted into a photon with desired frequency and bandwidth using the intracavity nonlinearity. As specific examples, we discuss a high-finesse, TE-TM double-mode photonic crystal cavity design that allows for direct generation of single photons at telecom wavelengths starting from an InAs/GaAs quantum dot with a 950 nm transition wavelength, and a scheme for direct optical coupling of such a quantum dot with a diamond nitrogen-vacancy center at 637 nm.

Nonlinear optical frequency conversion is widely used in fields as diverse as ultrahigh-resolution imaging [1] and telecommunications, as it allows for the generation of light in parts of the spectrum for which there are no convenient sources. Common implementations include optical parametric oscillators to make tunable femtosecond lasers in the infrared, and conversion of the 1064 nm Nd:YAG laser to make green laser sources via second-harmonic generation. Recently, nonlinear photonic crystal cavities [2, 3, 4, 5, 6] have emerged as promising systems in which similar nonlinear functionalities can be achieved at micron scales, which would enable the miniaturization of optical devices onto integrated platforms. While the majority of such work focuses on conversion of classical fields, these systems are now being applied to quantum optics and quantum information science [7, 8, 9, 10].

In recent years, there has also been a concerted research effort to develop on-demand single-photon sources using single quantum emitters strongly coupled to resonant optical microcavities (cavity QED) [11, 12, 13]. The strong coupling of the emitter to a resonant cavity results in preferential emission into the cavity mode of a single photon with frequency near the atomic resonance. Connecting pairs of such systems would form the basis for distributed quantum networks, where the emitters serve as processors and photons carry information between the nodes [14]. In practice, however, the photon emission occurs at wavelengths determined by the atomic resonance frequency. This is impractical, as it does not exploit the low-loss telecom frequency band for long-distance transmission and requires all emitters in a quantum network to be identical.

Here, we describe a novel approach to generate single photons with controllable wavelength and bandwidth. Our approach makes use of an integrated nonlinear optical cavity in which optical emission is directly frequency-shifted into the desired domain using intracavity nonlinear optical processes. This cavity-based generation technique is quite robust in that the maximum efficiency does not depend on an explicit phase-matching condition [15], as would occur in an extended nonlinear crystal or fiber, but rather only on the ratio of the cavity quality factor to mode volume (Q/V). As an example, we demonstrate a novel double mode TE-TM cavity design in a GaAs photonic crystal that is well-suited for the conversion of photons from quantum dots to the telecom band. We also present a similar GaP-based design for direct coupling between a nitrogen-vacancy center in diamond [16, 17, 18, 19] and an InAs/GaAs quantum dot [20, 21, 22], which could enable practical realization of a heterogeneous quantum network. In addition to effective wavelength control of single photons [7, 8, 9, 10], our approach enables the manipulation of their bandwidth, which is important for fast communication.

*These authors contributed equally to this work.; Electronic address: murray@seas.harvard.edu

The concept of single-photon spectral control

We first discuss the general protocol for generating single photons on demand at arbitrary frequencies using a nonlinear double-mode cavity, and introduce a simple theoretical model to derive the efficiency of the process. The system of interest is illustrated schematically in Fig. 1. As in standard cavity-based single-photon generation protocols [13, 14], a single three-level atom (or any other quantum dipole emitter) is resonantly coupled to one mode (here denoted a) of an optical cavity. The emitter is initialized in metastable state $|s\rangle$, and an external laser field with controllable Rabi frequency $\Omega(t)$ couples $|s\rangle$ to excited state $|e\rangle$. The transition between $|e\rangle$ and ground state $|g\rangle$ is resonantly coupled to cavity mode a (frequency ω_a), with a single-photon Rabi frequency g_1 . The relevant decay mechanisms (illustrated with gray arrows in the figure) are a leakage rate κ_a for photons to leave cavity mode a , and a rate γ that $|e\rangle$ spontaneously emits into free space rather than into the cavity. Conventionally, in absence of an optical nonlinearity, the control field $\Omega(t)$ creates a single atomic excitation at some desired time in the system, which via the coupling g_1 is converted into a single, resonant cavity photon. This photon eventually leaks out of the cavity and constitutes an outgoing, resonant single photon generated on demand whose spatial wave-packet can be shaped by properly choosing $\Omega(t)$ [14].

In our system, the cavity is also assumed to possess a second mode c with frequency ω_c , and our goal is to induce the single photon to exit at this frequency rather than ω_a . This can be achieved, provided that the cavity medium itself possesses a second-order ($\chi^{(2)}$) nonlinearity, by applying a classical pump field to the system at the difference frequency $\omega_b = \omega_a - \omega_c$. The induced coherent coupling rate between modes a and c is denoted g_2 . The field b need not correspond to a cavity mode. Mode c has a photon leakage rate, which we separate into an “inherent” rate, $\kappa_{c,in}$, and a “desirable” (extrinsic) rate, $\kappa_{c,ex}$. $\kappa_{c,in}$ characterizes the natural leakage into radiation modes and also absorption losses, and can be expressed in terms of the (unloaded) cavity quality factor as $\kappa_{c,in} = \omega_c/2Q_c$. $\kappa_{c,ex}$ characterizes the out-coupling rate into any external waveguide used for photon extraction. The total leakage of mode c is then $\kappa_c = \kappa_{c,in} + \kappa_{c,ex}$.

More quantitatively, the effective Hamiltonian for the system (in a rotating frame) is given by

$$\begin{aligned} H_I &= H_c + H_{loss}, \\ H_c &= \hbar g_1 (\sigma_{eg} a_a + \sigma_{ge} a_a^\dagger) + \hbar \Omega(t) (\sigma_{es} + \sigma_{se}) + \hbar g_2 (a_a^\dagger a_c + a_a a_c^\dagger), \\ H_{loss} &= -\frac{i\gamma}{2} \sigma_{ee} - \frac{i\kappa_a}{2} a_a^\dagger a_a - \frac{i(\kappa_{c,ex} + \kappa_{c,in})}{2} a_c^\dagger a_c, \end{aligned} \quad (1)$$

where H_c describes the coherent part of the system evolution (for simplicity we take $g_{1,2}, \Omega$ to be real), and H_{loss} is a non-Hermitian term characterizing the losses. $\sigma_{ij} = |i\rangle\langle j|$ are atomic operators, while a_i is the photon annihilation

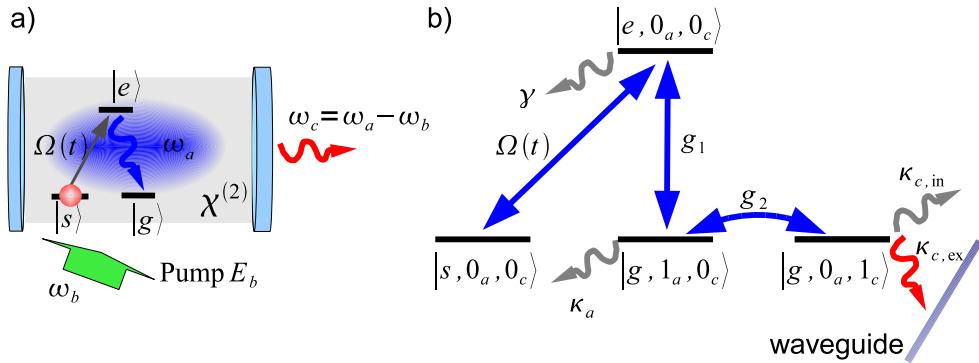


FIG. 1: **Schematic of single-photon frequency conversion** a) A single three-level emitter is coupled to a double-mode cavity that possesses a $\chi^{(2)}$ nonlinearity. After excitation, the emitter emits a photon into the cavity at frequency ω_a . When the cavity is irradiated by the pump beam at ω_b , the photon is converted to a second cavity mode at frequency ω_c . b) Level diagram: coherent coupling strengths are indicated with blue arrows, while gray arrows denote undesirable loss mechanisms. The emitter is controllably pumped from initial state $|s\rangle$ via an external laser field $\Omega(t)$ to excited state $|e\rangle$. The excited state $|e\rangle$ can reversibly emit a single photon into cavity mode a (while bringing the atom into state $|g\rangle$) at a rate g_1 , and can also decay into free space at rate γ . Mode a has an inherent decay rate given by κ_a . The nonlinearity allows the photon in mode a to be converted to one in mode c at a rate g_2 when the cavity is pumped by a laser of frequency $\omega_b = \omega_a - \omega_c$. The leakage rate of the frequency-converted photon at ω_c is split up into undesirable channels ($\kappa_{c,in}$) and desirable out-coupling to a nearby waveguide ($\kappa_{c,ex}$).

operator for mode i . The vacuum Rabi splitting g_1 can be written in the form $g_1 = \mathbf{d} \cdot \mathbf{E}_a(\mathbf{r})/\hbar$, where \mathbf{d} is the dipole matrix element of the $|g\rangle$ - $|e\rangle$ transition, and $\mathbf{E}_a(\mathbf{r})$ is the electric field amplitude per photon at the emitter position \mathbf{r} . The electric field per photon in mode $i = a, c$ is determined by the normalization

$$\frac{\hbar\omega_i}{2} = \int d\mathbf{r} \epsilon_0 \epsilon(\mathbf{r}) |\mathbf{E}_i(\mathbf{r})|^2, \quad (2)$$

where $\epsilon(\mathbf{r})$ is the dimensionless electric permittivity of the material. The nonlinearity parameter is given by [3]

$$g_2 = -\frac{\epsilon_0}{\hbar} \int d\mathbf{r} \chi_{ijk}^{(2)} E_{a,i}^* (E_{b,j} E_{c,k} + E_{c,j} E_{b,k}). \quad (3)$$

The amplitudes $E_{a,c}$ appearing above are normalized by Eq. (2), while E_b is the classical pump amplitude. Importantly, one can compensate for a small nonlinear susceptibility $\chi^{(2)}$ or field overlap (phase matching) simply by using larger pump amplitudes E_b to achieve a desired g_2 strength.

For a system initialized in $|s\rangle$, there can never be more than one excitation, and the system generally exists as a superposition of having the system in state $|s\rangle$ or $|e\rangle$ (with no photons) or having a photon in one of the modes a, c (and the emitter in $|g\rangle$),

$$|\psi(t)\rangle = c_s(t)|s\rangle + c_e(t)|e\rangle + c_a(t)|1_a\rangle + c_c(t)|1_c\rangle. \quad (4)$$

The system is initialized to $c_s(0) = 1$ with all other $c_i(0) = 0$ and the time evolution is given by $\dot{c}_j = -(i/\hbar)\langle j|H_I|\psi(t)\rangle$. In this effective wave-function approach, provided that $|s\rangle$ is always driven, $\sum_j |c_j|^2 \rightarrow 0$ as $t \rightarrow \infty$ due to losses, which can be connected with population leakage out of one of the aforementioned decay channels. In the limit that $\Omega(t)$ is small and varies slowly, all other $c_i(t)$ adiabatically follow $c_s(t)$ (see the Methods section), and one finds

$$\dot{c}_e(t) \approx -i\Omega(t)c_s(t) - \frac{1}{2} \left(\gamma + \frac{4g_1^2}{\kappa_a + 4g_2^2/\kappa_c} \right) c_e. \quad (5)$$

Physically, we can identify $\gamma_{\text{total}} = \gamma + \frac{4g_1^2}{\kappa_a + 4g_2^2/\kappa_c}$ as the cavity-enhanced total decay rate of $|e\rangle$, where the first (second) term corresponds to direct radiative emission (emission into mode a). Similarly, the denominator $\kappa_a + 4g_2^2/\kappa_c$ corresponds to the new total decay rate of mode a in the presence of an optical nonlinearity, as it yields a new channel for photons to effectively “decay” out of mode a into c at rate $4g_2^2/\kappa_c$. It is clear that some optimal value of g_2 exists for frequency conversion to occur. In particular, for no nonlinearity ($g_2 = 0$) this probability is non-existent. On the other hand, for $g_2 \rightarrow \infty$, one finds $\gamma_{\text{total}} = \gamma$, which indicates that the leakage from mode a into c is so strong that no cavity-enhanced emission occurs. Note that the use of time-varying control and pump fields allows for arbitrary shaping of the outgoing single-photon wavepacket at frequency ω_c , provided only that the photon bandwidth is smaller than κ_c (physically, the photon cannot leave faster than the rate determined by the cavity decay, see Methods). This feature is particularly useful in two respects. First, in practice κ_c can be much larger than γ , which enables extremely fast operation times. Second, pulse shaping is useful for constructing quantum networks, as it allows one to impedance-match the outgoing photon to other nodes of the network.

Based on the above arguments, the probability that a single photon of frequency ω_c is produced and extracted into the desired out-coupling waveguide is given by

$$F = \frac{C_{in}}{1 + \phi + C_{in}} \frac{\phi}{1 + \phi} \frac{\kappa_{c,ex}}{\kappa_c}, \quad (6)$$

where $\phi = 4g_2^2/(\kappa_a \kappa_c)$ characterizes the branching ratio in mode a of nonlinearity-induced leakage to inherent losses, and $C_{in} = 4g_1^2/\gamma \kappa_a$ is the inherent cavity cooperativity parameter for mode a in absence of nonlinearity. The first term on the right denotes the probability for $|e\rangle$ to decay into mode a , the second term the probability that a photon in mode a couples into mode c , and the third term the probability that a photon in mode c out-couples into the desired channel (see Methods for an exact calculation). ϕ depends on the pump amplitude E_b , with the optimal value $\phi = \sqrt{1 + C_{in}}$ yielding the maximum in F . For large $C_{in} \gg 1$, the maximum probability is

$$F \approx \left(1 - \frac{2}{\sqrt{C_{in}}} \right) \frac{\kappa_{c,ex}}{\kappa_c}. \quad (7)$$

Considering an emitter placed near the field maximum of mode a , $C_{in} \sim \frac{3Q_a}{2\pi^2} \frac{\lambda^3}{n_a^3 V_a} \frac{\gamma_0}{\gamma}$, where Q_a, V_a are the mode quality factor and volume, respectively, and n is the index of refraction at frequency ω_a . The ratio γ/γ_0 is the spontaneous

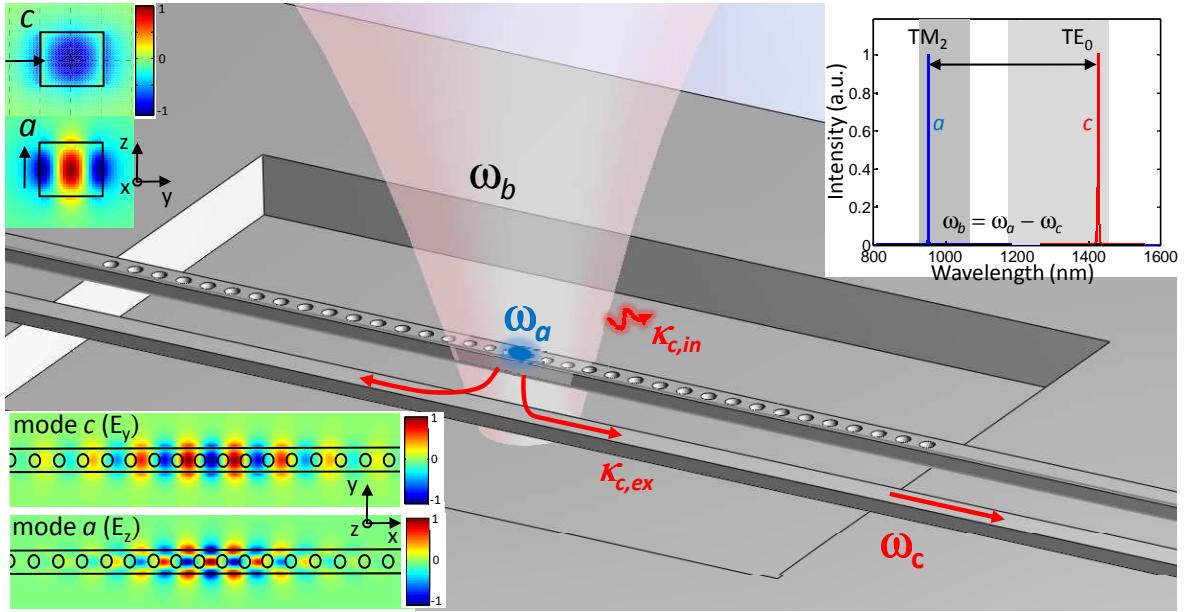


FIG. 2: Cavity mode characteristics Frequency conversion platform based on a photonic crystal nanobeam cavity, integrated extraction waveguide, and off-chip coupling laser (ω_b) tuned to the difference frequency of the modes. The cavity is formed by introducing a local perturbation into a periodic 1D line of air holes in the free-standing nanobeam. The desirable ($\kappa_{c,ex}$) and inherent ($\kappa_{c,in}$) loss channels from mode c are shown. The insets show the schematic cavity spectrum with photonic stopbands shown in grey, and the dominant field components of the TE_0 (ω_c) and TM_2 (ω_a) modes. The yz -plane cross-sections of the modes (upper left) show the E_y (E_z) component of mode c (a) at the center of the cavity, highlighting the mode overlap and polarizations. In the optimized structure (described in Methods), the TE mode at 1425 nm has $Q = 1.2 \times 10^7$ and $V_n = 0.77$, and the TM mode at 950 nm has $Q = 7.3 \times 10^4$ and $V_n = 1.45$ (V_n is the mode volume normalized by $(\lambda/n)^3$). The inherent peak cooperativities for the modes are $C_{in}^{TE} = 2.4 \times 10^7$ and $C_{in}^{TM} = 3.7 \times 10^4$, which are well into the strong coupling regime, as given by $C > 1$.

emission rate into non-cavity modes normalized by the spontaneous emission rate $\gamma_0 = n\omega_a^3|\mathbf{d}|^2/(3\pi\epsilon_0\hbar c^3)$ of an emitter embedded in an isotropic medium of index n . This ratio is expected to be of order 1 – 10 for our devices of interest, and thus the efficiency essentially depends only on Q_a/V_a .

Finally, while we have focused on the case of single-photon generation here, the reverse process can also be considered, where a single incoming photon at frequency ω_c is incident upon the system, converted into a photon in mode a , and coherently absorbed by an atom with the aid of an impedance-matched pulse $\Omega(t)$, causing its internal state to flip from $|g\rangle$ to $|s\rangle$. Generally, by time-reversal arguments [23], it can be shown that the probability F for single-photon storage is the same as that for generation.

Realization in a nonlinear photonic crystal cavity

In order to implement this frequency conversion scheme in a practical fashion, there are several constraints on the design of the cavity modes. For the nonlinear process to be efficient, mode a must have a high cooperativity (Q/V) to ensure strong coupling of the emitter (see Fig. 1). For mode c , a high Q factor (small κ_c) is important to maximize the nonlinear coupling parameter, ϕ , and hence reduce the pump power needed in order to reach the optimum nonlinear coupling strength, g_2 . The cavity should also be composed of a $\chi^{(2)}$ nonlinear material that is transparent in the desired frequency range. Finally, in order for the modes to couple efficiently via the nonlinear susceptibility of the cavity, they must have a large spatial overlap and the appropriate vector orientation, as determined by the elements of the $\chi^{(2)}$ tensor of the cavity material (see Eq. (3)).

As a host platform for the nonlinear cavity, the III-V semiconductors are promising candidates because of their significant second-order nonlinear susceptibilities and mature nanofabrication technologies. However, the symmetry of the III-V group $\chi^{(2)}$ tensor ($\chi_{ijk}^{(2)} \neq 0, i \neq j \neq k$) requires that the dominant field components of the modes be orthogonal in order to maximize the nonlinear coupling. It further implies that if the classical field which drives the nonlinear polarization is incident from the normal direction (e.g., from an off-chip laser), one of the cavity modes must have a TM polarization.

We adopt a photonic crystal platform to realize a wavelength-scale nonlinear cavity that meets these requirements. Recently, 2D photonic crystal nanocavities have shown great promise for strongly coupling an optical mode to a quantum dot emitter [21, 22]. In addition, they have been used as platforms for classical nonlinear optical generation and switching [2, 24]. The challenge, however, is to design a nonlinear photonic crystal nanocavity which supports two orthogonal, high cooperativity modes with a large mode field overlap.

To enable a monolithic cavity design which supports *both* TE and TM modes, we design a photonic crystal “nanobeam” cavity – a free-standing ridge waveguide patterned with a one-dimensional (1D) lattice of holes – for which we can control both TE and TM photonic bandstructures. Recently, there has been much interest in photonic crystal nanobeam cavities [25, 26, 27, 28, 29] due to their exceptional cavity figures of merit (Q and V), relative ease of design and fabrication, and potential as a platform to realize novel optomechanical effects [30, 31]. Our frequency conversion scheme can be realized in a similar structure, as shown in Fig. 2. We optimize two high cooperativity cavity modes by exploiting the different quasi-1D TE and TM photonic stopbands of the patterned nanobeam (shaded regions in the inset of Fig. 2). A key design point is that the TE and TM bandstructures can be tuned somewhat independently by varying the cross-sectional aspect ratio of the ridge. For example, in a nanobeam with a square cross-section, the two stopbands overlap completely. As the width-to-depth ratio of the waveguide is increased, the effective index of the TE modes increases relative to the TM modes, shifting the TE stopband to longer wavelengths.

Example implementations

As a first example, we design a GaAs photonic crystal nanobeam cavity with modes at 950 nm and 1425 nm suitable for directly generating single photons at telecom wavelengths from InAs/GaAs quantum dots. To achieve such a large spectral separation, we couple the fundamental TE₀ cavity mode to a *higher-order* TM₂ cavity mode (see inset Fig. 2). Crucially, the photonic crystal lattice tapering [27, 28, 31] is effective in enhancing the Q factors of *both TE and TM modes*. Details of the cavity parameters and optimization are provided in the Methods section. For this cavity, the coupling field (E_b) must have a wavelength $\lambda_b = 2.85 \mu\text{m}$ in order to efficiently drive the difference-frequency process. GaAs is an attractive nonlinear cavity material because it has a reasonably large $\chi^{(2)}$ strength [32], a high refractive index, and mature microfabrication techniques.

As evident in Fig. 2, the overlap of the two modes changes sign near the edges of the ridge compared to the middle due to the different symmetries of the TE₀ and TM₂ modes. However, the induced nonlinear polarization is dominated by the negatively signed anti-nodes near the middle of the ridge, and the imperfect overlap in the integral can be completely compensated for by a stronger pump beam. Thus, by selecting a higher order TM₂ mode, we have gained a larger frequency conversion bandwidth at the expense of the somewhat higher pump power required to overcome the ensuing phase mismatch. Note that the fundamental TE₀-TM₀ mode overlap is nearly ideal, and would be appropriate for applications requiring relatively small frequency shifts.

We now calculate the probability to convert a single photon from 950 to 1425 nm in our system. The optimized cavity design (see Methods) simultaneously yields high quality factors and small mode volumes, which allows for extremely high cooperativities for each mode ($C_{in} > 10^4$). From Eq. (7) we find that this enables an internal conversion probability of up to $F = 0.99$ when waveguide extraction efficiency is not taken into account. In practice, to efficiently out-couple the frequency-converted single photon into a waveguide, we require the ratio $\delta = \kappa_{c,ex}/\kappa_{c,in}$ to be large (i.e. overcoupled). The branching parameter ϕ scales as $P_b/(\kappa_{c,in}(1+\delta))$, and so to increase the extraction ratio δ , the pump power (P_b) must also be increased to maintain the optimal ϕ . Essentially, achieving good extraction efficiency requires one to intentionally increase the losses in mode c (via the out-coupling waveguide), which in turn requires more pump power to maintain the critical coupling. This relationship is made clear in Fig. 3, which plots the probability F as a function of pump power P_b and extraction ratio δ . For a given δ , the power P_b can be chosen to maximize the probability, reflecting the optimal value of g_2 for frequency conversion. The probability rises rapidly with P_b , reaching a maximum at relatively low powers (visible as the sharp ridge in the contours). Three fixed δ contours are plotted in Fig. 3(b), demonstrating that efficient extraction of frequency-converted photons can be realized at modest pumping powers. For example, for $\delta = 10$, an extraction probability of 0.7 can be realized with a coupling laser power of 3 mW focused in a diffraction-limited focal spot. We note that the absorption of GaAs at $2.85 \mu\text{m}$ is negligible, and so there will be no pump-induced heating. For this particular cavity design, the outgoing converted photon can be shaped to have a bandwidth of up to $\kappa_c \sim 100 \text{ MHz}$.

By exploiting the scaling properties of Maxwell’s equations, it is straightforward to design a similar cavity in GaP which supports modes at 637 nm and 950 nm. GaP is a nonlinear wide bandgap semiconductor which is transparent at 637 nm and has recently shown promise for the microcavity enhancement of diamond NV emission [33, 34]. Accounting for the exact refractive index dispersion and $\chi^{(2)}$ strength of GaP, we calculate that the internal frequency conversion probability is 0.99, and the extraction probability is 0.7 for $< 4 \text{ mW}$ coupling power. The 637-950 nm span would be sufficient to couple any pair of the most relevant quantum emitters, namely NV centers in diamond; atoms such as

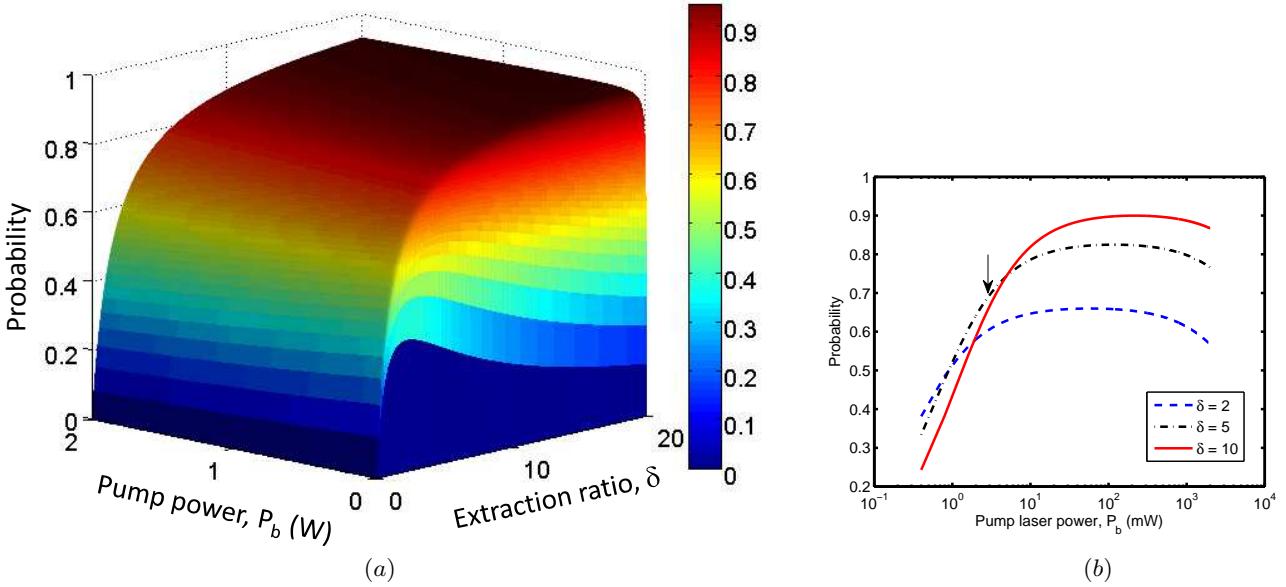


FIG. 3: **Probability of single-photon frequency conversion** from 950 nm to 1425 nm. The photon is coupled into a well-defined output channel at rate $\kappa_{c,ex}$. Note that the internal probability of conversion in the absence of an over-coupled extraction channel is 0.99. (a) Probability as a function of the pump laser power, P_b , and the extraction ratio, $\delta = \kappa_{c,ex}/\kappa_{c,in}$. For a given δ , there is an optimal operating power, P_b , as visible by the sharp contour ridge at small P_b . (b) Probability as a function of P_b for different values of δ . Because of the rapid rise in probability at low P_b , the system does not need to be operated at the optimum to achieve high conversion probabilities. For example, for $\delta = 10$, a probability of $F = 0.7$ can be achieved with a pump power $P_b = 3$ mW (indicated by the arrow).

Cs or Rb; and InAs/GaAs quantum dots. Such a cavity could also be integral to creating a stable, room temperature single-photon source emitting in the telecom band based on frequency-converted NV center emission in diamond [35]. Given that it may be difficult to span the large spectrum from 637 nm to telecom wavelengths in a single monolithic design, a 637-950 nm cavity could be the first stage of a two-step frequency conversion process involving our first example as the second stage. More generally, cascading allows our design to be extended to cover virtually any frequency span.

Outlook

We have shown that high-fidelity, intra-cavity frequency conversion of single photons from a dipole-like emitter can be achieved using a two-mode nonlinear cavity pumped by a classical field. Our general framework is valid for conversion between arbitrary frequencies, and the efficiency depends only on the cavity parameter Q/V . As realistic implementations, we propose two different high-cooperativity, double-mode photonic crystal nanocavities to enable highly efficient coupling from 950 nm – 1425 nm and 637 nm – 950 nm, respectively. Single-photon conversion between these wavelengths would allow, in the first instance, coupling of InAs/GaAs quantum dot emission [20, 21, 22] into low-loss optical fibers in the telecom spectrum. The second example would facilitate a direct optical connection between two types of solid-state emitters currently of great interest: a nitrogen-vacancy center in diamond [16, 17, 18, 19] and an InAs/GaAs quantum dot. Integrated together, the two designs could allow for cascaded frequency conversion of NV center emission into the telecom band. Further design improvements should lead to larger frequency spans and also lower pump power requirements (*e.g.*, by allowing ω_b to correspond to a third cavity mode). Although we have emphasized large frequency shifts in this paper, a smaller shift could be readily achieved by coupling the TE_0 mode with the fundamental TM_0 mode, which has a larger Q factor than the TM_2 mode studied here. The TE_0 - TM_0 modes have a larger spatial overlap, reducing the coupling power required for high probability frequency conversion.

Beyond the aforementioned applications, the techniques described here can potentially be extended to open up many intriguing opportunities. For example, the photon emission of a particular emitter could be shifted into wavelengths where high-efficiency detectors are available. It also allows coupling of atomic emitters such as Cs or Rb with solid-state emitters to create hybrid atom-photonic chips [36]. In addition, a number of quantum entanglement schemes for atoms rely on joint photon emission and subsequent detection to probabilistically project the atomic system into an entangled state [37, 38, 39]. Such schemes rely on the indistinguishability of photons emitted from each atom, and

implementing such techniques in nonlinear cavities could allow entanglement between different types of emitters. In addition, the protocol described here could be extended for generating narrow-bandwidth, entangled photon pairs with high efficiency and repetition rates, which are a valuable resource for applications such as quantum cryptography [40]. Our scheme could also be applicable in active materials, where laser wavelengths could be converted from easily accessible regions like 1500 nm to the mid-infrared range. Finally, it would be intriguing to combine these ideas with cavities that exhibit opto-mechanical coupling [31], which would potentially allow the photonic frequencies to be dynamically and rapidly tuned.

Methods

Derivation of nonlinear conversion efficiency

The state amplitudes of the wave-function given in Eq. (4) evolve under the interaction Hamiltonian H_I of Eq. (1) through the following equations,

$$\begin{aligned}\dot{c}_s &= -i\Omega(t)c_e, \\ \dot{c}_e &= -i\Omega(t)c_s - ig_1c_a - (\gamma/2)c_e, \\ \dot{c}_a &= -ig_1c_e - ig_2c_c - (\kappa_a/2)c_a, \\ \dot{c}_c &= -ig_2c_a - (\kappa_c/2)c_c.\end{aligned}\tag{8}$$

These equations describe both coherent evolution (terms proportional to $\Omega(t), g_{1,2}$) and population loss in the system (terms proportional to $\gamma, \kappa_{a,c}$). The population loss in the system can be connected to direct radiative emission of the excited state $|e\rangle$ (at a rate $\gamma|c_e|^2$), radiation leakage and absorption losses of mode a ($\kappa_a|c_a|^2$), and absorption and leakage out of mode c ($\kappa_c|c_c|^2$, of which $\kappa_{c,ex}|c_c|^2$ is successfully out-coupled to a waveguide). In general the efficiency of extracting a single photon of frequency ω_c out into the waveguide is thus

$$F = \frac{\int_0^\infty dt \kappa_{c,ex}|c_c(t)|^2}{\int_0^\infty dt \kappa_c|c_c(t)|^2 + \kappa_a|c_a(t)|^2 + \gamma|c_e(t)|^2}.\tag{9}$$

For arbitrary $\Omega(t)$, Eqs. (8) and (9) can be evaluated numerically. However, in certain limits one can find approximate solutions. In particular, when $\Omega(t)$ and its rate of change are small compared to the natural oscillation and decay rates of the system, the state amplitudes $c_{a,c,e}$ will follow the instantaneous value of $c_s(t)$. Formally, we can adiabatically eliminate these states, setting $\dot{c}_i = 0$ for $i = a, c, e$. Then, one finds

$$\dot{c}_s(t) = -\frac{2\Omega(t)^2}{\gamma_{\text{total}}}c_s(t),\tag{10}$$

while the other $c_i \propto c_s(t)$, with the proportionality coefficients being functions of $g_1, g_2, \kappa_a, \kappa_c, \Omega(t)$. The resulting substitution of the solutions of $c_i(t)$ into Eq. (9) allows great simplification because the integrands now become time-independent, and after some simplification yields Eq. (6). Self-consistency of the adiabatic elimination solution requires that the effective rate of population loss $\sim 4\Omega(t)^2/\gamma_{\text{total}}$ predicted from state $|s\rangle$ does not exceed the rate κ_c that a photon can leak out through the cavity mode c .

In the effective wave-function approach used here, the population leakage out of mode c can also be explicitly related to the shape of the outgoing single-photon wavepacket. For instance, we can model the linear coupling of cavity mode c to photons propagating in a single direction in a waveguide with the following Hamiltonian (in a rotating frame),

$$H_w = \int dk \hbar v(k - \omega_c/v) \hat{a}_k^\dagger \hat{a}_k - \hbar g_w \int dk (\hat{a}_c^\dagger \hat{a}_k e^{ikz_c} + h.c.).\tag{11}$$

Here k denotes the set of wavevectors of the continuum of waveguide modes, v is the velocity of waveguide fields, g_w is the coupling strength between cavity and waveguide modes, and z_c denotes the position along the waveguide where the cavity is coupled to it (for simplicity we set $z_c = 0$ from this point on). Since we are now explicitly accounting for the waveguide degrees of freedom, we add a term $\int dk c_k(t)|1_k\rangle$ to the effective wave-function of the system. The equations of motion of the total system are identical to Eq. (8), except that

$$\dot{c}_c = -ig_2c_a - (\kappa_{c,in}/2)c_c + ig_w \int dk c_k,\tag{12}$$

$$\dot{c}_k = -iv(\delta k)c_k + ig_w c_c,\tag{13}$$

where $\delta k = k - \omega_c/v$. Compared to Eq. (8), we have now included the coupling of mode c to the waveguide, and accordingly have replaced $\kappa_c \rightarrow \kappa_{c,in}$ in the equation for \dot{c}_c since the leakage into the waveguide should be accounted for by the new coupling terms. The equation for \dot{c}_k can be formally integrated; assuming that the waveguide initially is unoccupied, $c_k(0) = 0$, one has

$$c_k(t) = ig_w \int_0^t dt' c_c(t') e^{-ic\delta k(t-t')}. \quad (14)$$

Substituting this into the equation for \dot{c}_c and performing the Wigner-Weisskopf approximation [41], one recovers the expression for \dot{c}_c in Eq. (8) by identifying $\kappa_{c,ex} = 2\pi g_w^2/v$. The one-photon wave-function [41] is given by $\psi_w(z, t) = \langle vac | \hat{E}_w(z, t) | \psi(0) \rangle = (\sqrt{2\pi} ig_w/v) \Theta(z) c_c(t - z/v)$, where $\Theta(z)$ is the step function. The wave-function shape is thus directly proportional to $c_c(t)$. Under adiabatic elimination,

$$\psi_w(z, t) = \frac{\sqrt{2\pi} ig_w}{v} \Theta(z) \frac{8ig_1g_2}{\gamma_{\text{total}}(\kappa_a\kappa_c + 4g_2^2)} \Omega(t - z/v) c_s(t - z/v), \quad (15)$$

and thus for a desired (and properly normalized) pulse shape ψ_w one needs only to solve Eqs. (15) and (10) to obtain the corresponding external field $\Omega(t)$. It is straightforward to show that the normalization is given by $\int dz |\psi_w(z, t \rightarrow \infty)|^2 = F$ provided that $c_s(\infty) \rightarrow 0$. This normalization reflects the probability that a single photon ends up in the waveguide.

Nonlinear cavity design

The nanobeam cavities are formed by a 4-period taper in the size and spacing of the holes in the uniform photonic “mirror” on both sides of the cavity center in order to introduce a localized potential for the TE and TM modes. The 950-1425 nm cavity nanobeam has width $w = 420$ nm and depth $d = 307.5$ nm, and the hole spacing tapers from $a_0 = 360$ nm in the mirror to $a_c = 337$ nm in the center. The holes were made elliptical to give an additional design parameter to separately optimize the TE and TM mode Q factors. The elliptical hole semi-axes are 84 nm and 108 nm in the mirror section, and the hole size-to-spacing ratio is held constant through the taper section. This design yields cavity parameters of $Q = 1.2 \times 10^7$ and $V_n = 0.77$ for the TE mode, and $Q = 7.3 \times 10^4$ and $V_n = 1.45$ for the TM mode (V_n is the mode volume normalized by $(\lambda/n)^3$). The factor $\gamma/\gamma_0 = 0.10$ (0.20) for the TE (TM) mode is determined by simulating the total power emitted by a non-resonant dipole source in the cavity center. We have accounted for the index dispersion of our candidate material, GaAs, for which $n(1425 \text{ nm}) = 3.38$ and $n(950 \text{ nm}) = 3.54$ [42].

The nonlinear parameter g_2 is determined by calculating the volume integral of Eq. (3) using the exact mode fields, E_a and E_c , extracted from our 3D-FDTD calculation. Because the mode fields are oriented along the y and z -axes, respectively, as defined in Fig. 2(c), the classical field which drives the difference frequency generation, E_b , must be polarized along x . This field has a frequency $\omega_b = \omega_a - \omega_c$, which corresponds to a wavelength $\lambda_b = 2.85 \mu\text{m}$. The relevant nonlinear susceptibility tensor elements are $\chi_{xyz}^{(2)}(\text{GaAs}) = 2d_{14} = 550 \text{ pm/V}$ and $\chi_{xyz}^{(2)}(\text{GaP}) = 320 \text{ pm/V}$ [32, 43].

We assume the classical field is constant over the spatial extent of the cavity modes, which allows $E_{b,x}$ to be taken in front of the integral for g_2 , giving

$$g_2 = -\frac{\epsilon_0 E_{b,x}}{\hbar} \int d\mathbf{r} \chi_{xyz}^{(2)} E_{a,y}^* E_{c,z}. \quad (16)$$

To justify this assumption, we simulated a Gaussian beam with $\lambda_b = 2.85 \mu\text{m}$ that is focused by a lens with a modest numerical aperture (NA) of 0.5 onto a ridge waveguide, and found that the average field amplitude is approximately uniform over the linear extent of our cavity modes (approx. $x = -1 \mu\text{m}$ to $+1 \mu\text{m}$). In the g_2 calculation, the magnitude of $E_{b,x}$ for a given beam power, P_b , is then determined from the relation $P_b = \epsilon_0 c \pi r^2 E_{b,x}^2 / 4$, where r is the focal spot radius.

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[1] P. J. Campagnola, M.-d. Wei, A. Lewis, and L. M. Loew, *Biophys. J.* **77**, 3341 (1999).

[2] M. W. McCutcheon, J. F. Young, G. W. Rieger, D. Dalacu, S. Frédéric, P. J. Poole, and R. L. Williams, *Phys. Rev. B* **76**, 245104 (2007).

[3] A. Rodriguez, M. Soljacic, J. D. Joannopoulos, and S. G. Johnson, *Optics Express* **15**, 7303 (2007).

[4] J. Bravo-Abad, A. Rodriguez, P. Bermel, S. G. Johnson, J. D. Joannopoulos, and M. Soljacic, *Optics Express* **15**, 16161 (2007).

[5] M. Liscidini and L. C. Andreani, *Phys. Rev. E* **73**, 016613 (2006).

[6] A. R. Cowan and J. F. Young, *Semicond. Sci. Technol.* **20**, R41 (2005).

[7] A. P. VanDevender and P. G. Kwiat, *J. Mod. Opt.* **51**, 1433 (2004).

[8] C. Langrock, E. Diamanti, R. V. Roussev, Y. Yamamoto, M. M. Fejer, and H. Takesue, *Opt. Lett.* **30**, 1725 (2005).

[9] S. Tanzilli, W. Tittel, M. Halder, O. Alibart, P. Baldi, N. Gisin, and H. Zbinden, *Nature* **437**, 116 (2005).

[10] A. P. VanDevender and P. G. Kwiat, *J. Opt. Soc. Am. B* **24**, 295 (2007).

[11] P. Michler, A. Kiraz, C. Becher, W. V. Schoenfeld, P. M. Petroff, L. Zhang, E. Hu, and A. Imamoglu, *Science* **290**, 2282 (2000).

[12] M. Pelton, C. Santori, J. Vučković, B. Zhang, G. S. Solomon, J. Plant, and Y. Yamamoto, *Phys. Rev. Lett.* **89**, 233602 (2002).

[13] J. McKeever, A. Boca, A. D. Boozer, R. Miller, J. R. Buck, A. Kuzmich, and H. J. Kimble, *Science* **303**, 1992 (2004).

[14] J. I. Cirac, P. Zoller, H. J. Kimble, and H. Mabuchi, *Phys. Rev. Lett.* **78**, 3221 (1997).

[15] R. W. Boyd, *Nonlinear Optics* (Academic, New York, 1992).

[16] M. V. G. Dutt, L. Childress, L. Jiang, E. Togan, J. Maze, F. Jelezko, A. S. Zibrov, P. R. Hemmer, and M. D. Lukin, *Science* **316**, 1312 (2007).

[17] C. Santori, P. Tamarat, P. Neumann, J. Wrachtrup, D. Fattal, R. G. Beausoleil, J. Rabeau, P. Olivero, A. D. Greentree, S. Prawer, et al., *Phys. Rev. Lett.* **97**, 247401 (2006).

[18] T. Gaebel, M. Domhan, I. Popa, C. Wittmann, P. Neumann, F. Jelezko, J. R. Rabeau, N. Stavrias, A. D. Greentree, S. Prawer, et al., *Nature Physics* **2**, 408 (2006).

[19] R. Hanson, F. M. Mendoza, R. J. Epstein, and D. D. Awschalom, *Phys. Rev. Lett.* **97**, 087601 (2006).

[20] K. Srinivasan and O. Painter, *Nature* **450**, 862 (2007).

[21] K. Hennessy, A. Badolato, M. Winger, D. Gerace, M. Atatüre, S. Gulde, S. Falt, E. L. Hu, and A. Imamoglu, *Nature* **445**, 896 (2007).

[22] D. Englund, A. Faraon, I. Fushman, N. Stoltz, P. Petroff, and J. Vučković, *Nature* **450**, 857 (2007).

[23] A. V. Gorshkov, A. André, M. Fleischhauer, A. S. Sørensen, and M. D. Lukin, *Phys. Rev. Lett.* **98**, 123601 (2007).

[24] T. Tanabe, M. Notomi, S. Mitsugi, A. Shinya, and E. Kuramochi, *Appl. Phys. Lett.* **87**, 151112 (2005).

[25] C. Sauvan, G. Lecamp, P. Lalanne, and J. Hugonin, *Opt. Express* **13**, 245 (2005).

[26] A. R. M. Zain, N. P. Johnson, M. Sorel, and R. M. D. la Rue, *Opt. Express* **16**, 12084 (2008).

[27] M. W. McCutcheon and M. Lončar, *Opt. Express* **16**, 19136 (2008).

[28] M. Notomi, E. Kuramochi, and H. Taniyama, *Opt. Express* **16**, 11095 (2008).

[29] P. Deotare, M. W. McCutcheon, I. W. Frank, M. Khan, and M. Loncar, arXiv:0901.4158v2 [physics.optics] (2009).

[30] M. L. Povinelli, M. Loncar, M. Ibanescu, E. J. Smythe, S. G. Johnson, F. Capasso, and J. D. Joannopoulos, *Opt. Lett.* **30**, 3042 (2005).

[31] M. Eichenfield, R. Camacho, J. Chan, K. J. Vahala, and O. Painter, arXiv:0812.2953v1 [physics.optics] (2008).

[32] S. Singh, *Nonlinear optical materials*, CRC Handbook of Laser Science and Technology, Vol. III: Optical Materials, Part I, vol. III (CRC Press, 1986).

[33] K.-M. C. Fu, C. Santori, P. E. Barclay, I. Aharonovich, S. Prawer, N. Meyer, A. M. Holm, and R. G. Beausoleil, *Appl. Phys. Lett.* **93**, 234107 (2008).

[34] K. Rivoire, A. Faraon, and J. Vučković, *Appl. Phys. Lett.* **93**, 063103 (2008).

[35] C. Kurtsiefer, S. Mayer, P. Zarda, and H. Weinfurter, *Phys. Rev. Lett.* **85**, 290 (2000).

[36] P. E. Barclay, K. Srinivasan, O. Painter, B. Lev, and H. Mabuchi, *Appl. Phys. Lett.* **13**, 801 (2005).

[37] C. Cabrillo, J. I. Cirac, P. García-Fernández, and P. Zoller, *Phys. Rev. A* **59**, 1025 (1999).

[38] S. Bose, P. L. Knight, M. B. Plenio, and V. Vedral, *Phys. Rev. Lett.* **83**, 5158 (1999).

[39] L.-M. Duan and H. J. Kimble, *Phys. Rev. Lett.* **90**, 253601 (2003).

[40] A. K. Ekert, *Phys. Rev. Lett.* **67**, 661 (1991).

[41] M. O. Scully and M. S. Zubairy, *Quantum Optics* (Cambridge University Press, Cambridge, UK, 1997).

[42] E. D. Palik, *Handbook of optical constants of solids*, vol. 1 (Academic Press, Inc., 1985).

[43] I. Shoji, T. Kondo, A. Kitamoto, M. Shirane, and R. Ito, *J. Opt. Soc. Am. B* **14**, 2268 (1997).